



10/698 110

*Handwritten signature/initials*

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*Handwritten signature of Frank C. Eisenschenk*

Frank C. Eisenschenk, Ph.D., Patent Attorney

REQUEST FOR CERTIFICATE OF  
CORRECTION UNDER 37 CFR 1.322  
AND UNDER 37 CFR 1.323

Docket No. UF-380

Patent No. 7,012,165

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants : William R. Dolbier, Jr., Buvaneswari Gopal  
Issued : March 14, 2006  
Patent No. : 7,012,165  
For : Oxidative Fluorination of Aromatic Derivatives by Copper (II) Fluoride and Silver (I) Fluoride

Mail Stop Certificate of Corrections Branch  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

**Certificate  
APR 26 2006  
of Correction**

REQUEST FOR CERTIFICATE OF CORRECTION  
UNDER 37 CFR 1.322 (OFFICE MISTAKE) AND  
UNDER 37 CFR 1.323 (APPLICANTS' MISTAKE)

Sir:

A Certificate of Correction (in duplicate) for the above-identified patent has been prepared and is attached hereto.

In the left-hand column below is the column and line number where errors occurred in the patent. In the right-hand column is the page and line number in the application where the correct information appears.

**Patent Reads:**

Title page, Item (74):

"& Salinwanchik"

**Application Reads:**

Payment Fee Sheet:

--& Saliwanchik--

| 04/24/2006 BABRAHA1 00000096 190065 7012165  
| 01 FC:1811 100.00 DA

**APR 27 2006**

Column 3, line 20:

"Hg<sub>2</sub>F<sub>2</sub>. As illustrated"

**Patent Reads:**

Column 3, line 29:

"fluorbenzene: diflorobenzene"

Column 4, line 41:

"flurobenzene"

**Patent Reads:**

Column 11, line 1:

"flourinate"

**Patent Reads:**

Column 11, line 3:

"fluoronation"

**Patent Reads:**

Column 11, line 4:

"flourobenzene"

**Patent Reads :**

Column 11, line 4:

"fluorotoluene"

Page 4, lines 19-21:

--Hg<sub>2</sub>F<sub>2</sub>.

As illustrated--

**Application Should Read:**

Page 4, line 26:

--fluorobenzene: diflorobenzene--

Page 5, line 27:

--fluorobenzene--

**Application Reads:**

Page 11, line 6:

--fluorinate--

**Application Should Read:**

Page 11, line 7:

--fluorination--

**Application Reads:**

Page 11, line 8:

--fluorobenzene--

**Application Should Read :**

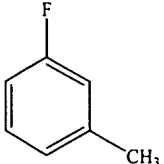
Page 11, line 8:

--fluorotoluene--

APR 27 2006

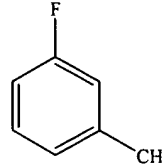
**Patent Reads:**Column 12, Lines 5-10, Table 9:

“

Reactants	Temperature °C	Product Yields
	350	

**Application Reads:**Page 12, Lines 3-4:

--

Reactants	Temperature °C	Product Yields
	350	only 2-4% fluorination

--

Column 15, Line 3:

“components a temperature”

Amendment dated April 13, 2005 (claim 20,  
Line 8):

--components at a temperature--.

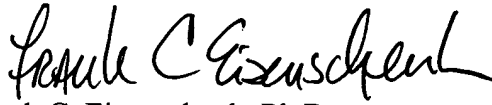
A true and correct copy of Fee Transmittal letter, Amendment dated April 13, 2005 and pages 4, 5, 11, and 12 of the specification as filed which support Applicants' assertion of the errors on the part of the Patent Office accompanies this Certificate of Correction.

The Commissioner is authorized to charge the fee of \$100.00 for the amendment to Deposit Account No. 19-0065. The Commissioner is also authorized to charge any additional fees as required under 37 CFR 1.20(a) to Deposit Account No. 19-0065. Two copies of this letter are enclosed for Deposit Account authorization.

APR 27 2006

Approval of the Certificate of Correction is respectfully requested.

Respectfully submitted,



Frank C. Eisenschenk, Ph.D.

Patent Attorney

Registration No. 45,332

Phone No.: 352-375-8100

Fax No.: 352-372-5800

Address: P.O. Box 142950  
Gainesville, FL 32614-2950

FCE/atn

Attachments: Copy of Fee Transmittal Letter, Amendment dated April 13, 2005 and pages 4, 5, 11, and 12 of the specification.

## PART B - FEE(S) TRANSMITTAL

Complete and send this form, together with applicable fee(s), to: Mail

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INSTRUCTIONS: This form should be used for transmitting the ISSUE FEE and PUBLICATION FEE (if required). Blocks 1 through 5 should be completed where appropriate. All further correspondence including the Patent, advance orders and notification of maintenance fees will be mailed to the current correspondence address as indicated unless corrected below or directed otherwise in Block 1, by (a) specifying a new correspondence address; and/or (b) indicating a separate "FEE ADDRESS" for maintenance fee notifications.

CURRENT CORRESPONDENCE ADDRESS (Note: Use Block 1 for any change of address)

23557 7590 06/24/2005

SALIWANCHIK LLOYD & SALIWANCHIK  
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PO BOX 142950  
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Attn: Jenna M. Morrison

Note: A certificate of mailing can only be used for domestic mailings of the Fee(s) Transmittal. This certificate cannot be used for any other accompanying papers. Each additional paper, such as an assignment or formal drawing, must have its own certificate of mailing or transmission.

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Gwendolyn L. Daniels (Depositor's name)  
*Gwendolyn L. Daniels* (Signature)  
July 27, 2005 (Date)

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
10/698,110	10/31/2003	William R. Dolbier JR.	UF-380	8916

TITLE OF INVENTION: ~~OXIDATIVE FLUORINATION OF AROMATIC DERIVATIVES BY COPPER (II) FLUORIDE AND SILVER (I) FLUORIDE~~  
Oxidative Fluorination of Aromatic and Chloroaromatic Derivatives (as amended)

APPLN. TYPE	SMALL ENTITY	ISSUE FEE	PUBLICATION FEE	TOTAL FEE(S) DUE	DATE DUE
nonprovisional	YES	\$700	\$300	\$1000	09/26/2005

EXAMINER	ART UNIT	CLASS-SUBCLASS
WITHERSPOON, SIKARL A	1621	570-127000

1. Change of correspondence address or indication of "Fee Address" (37 CFR 1.363).  
☐ Change of correspondence address (or Change of Correspondence Address form PTO/SB/122) attached.  
☐ "Fee Address" indication (or "Fee Address" Indication form PTO/SB/47; Rev 03-02 or more recent) attached. Use of a Customer Number is required.

2. For printing on the patent front page, list  
(1) the names of up to 3 registered patent attorneys or agents OR, alternatively,  
(2) the name of a single firm (having as a member a registered attorney or agent) and the names of up to 2 registered patent attorneys or agents. If no name is listed, no name will be printed.

Saliwanchik, Lloyd  
& Saliwanchik  
\_\_\_\_\_  
\_\_\_\_\_  
\_\_\_\_\_

## 3. ASSIGNEE NAME AND RESIDENCE DATA TO BE PRINTED ON THE PATENT (print or type)

PLEASE NOTE: Unless an assignee is identified below, no assignee data will appear on the patent. If an assignee is identified below, the document has been filed for recordation as set forth in 37 CFR 3.11. Completion of this form is NOT a substitute for filing an assignment.

(A) NAME OF ASSIGNEE

University of Florida

(B) RESIDENCE: (CITY and STATE OR COUNTRY)

Gainesville, FL

Please check the appropriate assignee category or categories (will not be printed on the patent): ☐ Individual ☐ Corporation or other private group entity ☐ Government

## 4a. The following fee(s) are enclosed:

- ☒ Issue Fee  
☒ Publication Fee (No small entity discount permitted)  
☒ Advance Order - # of Copies 10

## 4b. Payment of Fee(s):

- ☐ A check in the amount of the fee(s) is enclosed.  
☐ Payment by credit card. Form PTO-2038 is attached.  
☒ The Director is hereby authorized by charge the required fee(s), or credit any overpayment, to Deposit Account Number 19-0065 (enclose an extra copy of this form).

## 5. Change in Entity Status (from status indicated above)

- ☐ a. Applicant claims SMALL ENTITY status. See 37 CFR 1.27. ☐ b. Applicant is no longer claiming SMALL ENTITY status. See 37 CFR 1.27(g)(2).

The Director of the USPTO is requested to apply the Issue Fee and Publication Fee (if any) or to re-apply any previously paid issue fee to the application identified above.  
NOTE: The Issue Fee and Publication Fee (if required) will not be accepted from anyone other than the applicant; a registered attorney or agent; or the assignee or other party in interest as shown by the records of the United States Patent and Trademark Office.

Authorized Signature *Jenna M. Morrison*  
Typed or printed name Jenna M. Morrison

Date July 27, 2005Registration No. 55,468

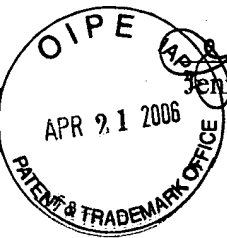
This collection of information is required by 37 CFR 1.311. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, Virginia 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, Virginia 22313-1450.

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APR 27 2006

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AMENDMENT UNDER 37 CFR §1.111  
Examining Group 1621  
Patent Application  
Docket No. UF-380  
Serial No. 10/698,110



Jenna M. Morrison  
Jenna M. Morrison, Patent Attorney

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Examiner : Sikarl A. Witherspoon  
Art Unit : 1621  
Applicants : William R. Dolbier, Jr., Buvaneswari Gopal  
Serial No. : 10/698,110  
Filed : October 31, 2003  
Conf. No. : 8916  
For : Oxidative Fluorination of Aromatic Derivatives by Copper (II) Fluoride and  
Silver (I) Fluoride

MAIL STOP AMENDMENT  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313

AMENDMENT UNDER 37 CFR §1.111

Sir:

In response to the Office Action dated January 13, 2005, please amend the above-identified  
patent application as follows:

**Amendments to the Specification begin on page 2 of this paper.**

**Amendments to the Claims follow the Specification Amendment Section of this paper.**

**Remarks/Arguments follow the Claims Amendment Section of this paper.**

APR 27 2006

In the Specification

Please replace the title ("Oxidative Fluorination of Aromatic Derivatives by Copper (II) Fluoride and Silver (I) Fluoride") of the application on page 1, line 3, with the new title as presented herein below:

Oxidative Fluorination of Aromatic and Chloroaromatic Derivatives

In the Claims

Claim 1 (Currently Amended): A method of fluorinating an aromatic compound or chloroaromatic compound comprising the steps of:

- a) mixing at least one active fluorinating agent selected from the group consisting of  $\text{CuF}_2$ ,  $\text{AgF}$ ,  $\text{HgF}_2$ ,  $\text{TeF}_4$ ,  $\text{MnF}_4$ ,  $\text{FeF}_3$ , and  $\text{CoF}_{2-4}$  with at least one support selected from the group consisting of activated carbon,  $\text{ZnF}_2$ ,  $\text{CaF}_2$ ,  $\text{MgF}_2$ ,  $\text{AlF}_3$ , and combinations of activated carbon,  $\text{ZnF}_2$ ,  $\text{CaF}_2$ ,  $\text{MgF}_2$ , or  $\text{AlF}_3$ ;
- b) heating said mixture ~~[[to at]]~~ a temperature of at least  $300^\circ\text{C}$  or  $350^\circ\text{C}$ ; and
- c) contacting said mixture with an aromatic compound, a chloroaromatic compound, a mixture of aromatic compounds, a mixture of chloroaromatic compounds, or a mixture of chloroaromatic and aromatic compounds.

Claim 2 (Original): The method according to claim 1, wherein said method further comprises recovering fluorinated aromatic or chloroaromatic compounds.

Claim 3 (Original): The method according to claim 1, wherein said temperature is at least  $400^\circ\text{C}$ .

Claim 4 (Original): The method according to claim 1, wherein said temperature is at least  $425^\circ\text{C}$ .

Claim 5 (Original): The method according to claim 1, wherein said temperature is at least  $450^\circ\text{C}$ .

Claim 6 (Original): The method according to claim 1, wherein said temperature is at least  $500^\circ\text{C}$ .



Claim 7 (Original): The method according to claim 1, wherein said aromatic or chloroaromatic compound is selected from the group consisting of benzene, chlorobenzene, substituted benzene, substituted chlorobenzene, pyridines, chloropyridines, substituted pyridines, substituted chloropyridines, naphthalene, substituted naphthalenes, chloronaphthalene, substituted chloronaphthalenes, toluene, chlorotoluene, substituted toluene, and substituted chlorotoluene.

Claim 8 (Original): The method according to claim 1, wherein aromatic compounds are contacted with said mixture.

Claim 9 (Original): The method according to claim 1, wherein chloroaromatic compounds are contacted with said mixture.

Claim 10 (Original): The method according to claim 1, wherein a mixture of chloroaromatic and aromatic compounds are contacted with said mixture.

Claim 11 (Original): The method according to claim 9, wherein said mixture comprises AgF and at least one support.

Claim 12 (Original): The method according to claim 8, wherein said aromatic compounds are aromatic hydrocarbons.

Claim 13 (Original): The method according to claim 12, wherein said aromatic compounds are contacted with a mixture comprising  $\text{CuF}_2$  and at least one support.

Claim 14 (Original): The method according to claim 9, wherein said chloroaromatic compound is ortho-dichlorobenzene, para-dichlorobenzene, meta-dichlorobenzene, a chloropyridine, chloronaphthalene, a chlorpyridine, chlorotoluene, substituted ortho-dichlorobenzene, substituted para-dichlorobenzene, substituted meta-dichlorobenzene, a substituted chloropyridine, substituted chloronaphthalene, a substituted chlorpyridine, substituted chlorotoluene, or mixtures thereof.

Claim 15 (Currently Amended): The method according to claim 14, wherein said ~~mixture~~ chloroaromatic compound is contacted by a mixture comprising AgF and at least one support.

Claim 16 (Canceled).

Claim 17 (Original): The method according to claim 12, wherein said aromatic compounds are contacted with a mixture consisting of  $\text{CuF}_2$  and at least one support.

Claim 18 (Original): The method according to claim 11, wherein said mixture further comprises  $\text{CuF}_2$ .

Claim 19 (Original): The method according to claim 13, wherein said mixture further comprises AgF.

Claim 20 (Currently Amended): A method of fluorinating an aromatic compound comprising combining an aromatic compound, a chloroaromatic compound, a mixture of aromatic compounds, a mixture of chloroaromatic compounds, or a mixture of chloroaromatic and aromatic compounds and a fluorinating composition comprising at least one active fluorinating agent selected from the group consisting of  $\text{CuF}_2$ , AgF,  $\text{HgF}_2$ ,  $\text{TeF}_4$ ,  $\text{MnF}_4$ ,  $\text{FeF}_3$ , and  $\text{CoF}_{2-4}$  and at least one support selected from the group consisting of activated carbon,  $\text{ZnF}_2$ ,  $\text{CaF}_2$ ,  $\text{MgF}_2$ ,  $\text{AlF}_3$ , and combinations of activated carbon,  $\text{ZnF}_2$ ,  $\text{CaF}_2$ ,  $\text{MgF}_2$ , or  $\text{AlF}_3$  and heating the combined components ~~[[to at]]~~ a temperature of at least  $350^\circ\text{C}$ .

Claim 21 (New): The method according to claim 5, wherein said mixture comprises one part active fluorinating agent and one part to three parts of the at least one support.

Claim 22 (New): The method according to claim 21, wherein the active fluorinating agent is  $\text{CuF}_2$  and at least one support is  $\text{AlF}_3$ .

Claim 23 (New): The method according to claim 5, wherein said mixture comprises one part active fluorinating agent and two parts of the at least one support.

Claim 24 (New): The method according to claim 23, wherein the active fluorinating agent is  $\text{CuF}_2$  and at least one support is  $\text{AlF}_3$ .

Claim 25 (New): The method according to claim 6, wherein said mixture comprises one part active fluorinating agent and one part to three parts of the at least one support.

Claim 26 (New): The method according to claim 6, wherein said mixture comprises one part active fluorinating agent and two parts of the at least one support.

Claim 27 (New): The method according to claim 25, wherein the active fluorinating agent is  $\text{CuF}_2$  and at least one support is  $\text{AlF}_3$ .

Claim 28 (New): The method according to claim 26, wherein the active fluorinating agent is  $\text{CuF}_2$  and at least one support is  $\text{AlF}_3$ .

Claim 29 (New): A method for producing fluorobenzene comprising:

- a) mixing at least one active fluorinating agent selected from the group consisting of  $\text{CuF}_2$ ,  $\text{AgF}$ ,  $\text{HgF}_2$ ,  $\text{TeF}_4$ ,  $\text{MnF}_4$ ,  $\text{FeF}_3$ , and  $\text{CoF}_{2-4}$  with at least one support selected from the group consisting of activated carbon,  $\text{ZnF}_2$ ,  $\text{CaF}_2$ ,  $\text{MgF}_2$ ,  $\text{AlF}_3$ , and combinations of activated carbon,  $\text{ZnF}_2$ ,  $\text{CaF}_2$ ,  $\text{MgF}_2$ , or  $\text{AlF}_3$ ;
- b) heating said mixture at a temperature of at least  $450^\circ\text{C}$ ; and
- c) contacting said mixture with an aromatic compound, a chloroaromatic compound, a mixture of aromatic compounds, a mixture of chloroaromatic compounds, or a mixture of chloroaromatic and aromatic compounds.

Claim 30 (New): The method according to claim 29, wherein the temperature is at least  $500^\circ\text{C}$ .

Claim 31 (New): The method according to claim 29, wherein said aromatic or chloroaromatic compound is selected from the group consisting of benzene, chlorobenzene, substituted benzene, substituted chlorobenzene, pyridines, chloropyridines, substituted pyridines, substituted chloropyridines, naphthalene, substituted naphthalenes, chloronaphthalene, substituted chloronaphthalenes, toluene, chlorotoluene, substituted toluene, and substituted chlorotoluene.

Claim 32 (New): The method according to claim 29, wherein said mixture comprises one part active fluorinating agent and one part to three parts of the at least one support.

Claim 33 (New): The method according to claim 32, wherein the active fluorinating agent is  $\text{CuF}_2$  and at least one support is  $\text{AlF}_3$ .

Claim 34 (New): The method according to claim 29, wherein said mixture comprises one part active fluorinating agent and two parts of the at least one support.

Claim 35 (New): The method according to claim 34, wherein the active fluorinating agent is  $\text{CuF}_2$  and at least one support is  $\text{AlF}_3$ .

Claim 36 (New): The method according to claim 33, wherein the support is  $\text{AlF}_3$ .

Claim 37 (New): The method according to claim 35, wherein the support is  $\text{AlF}_3$ .

Claim 38 (New): The method according to claim 29, wherein said method further comprises recovering fluorinated aromatic or chloroaromatic compounds.

Remarks

Claims 1-20 were pending in the subject application. By this Amendment, Applicants have amended the title of the application such that it is more clearly indicative of the invention to which the claims are directed. Claims 1, 15, and 20 have been amended, support for which can be found throughout the specification including page 2, lines 13-31; page 5, lines 1-9; and Example 5, and claim 16 has been canceled. New claims 21- 38 have been added, support for which can be found on, for example, page 2, lines 5-31; page 5, lines 16-17; page 6, Tables 1-2, page 7, Table 3; and page 8, lines 5-17. Thus, claims 1-15 and 17-38 are presently before the examiner.

Claims 1-20 are rejected under 35 U.S.C. § 103(a) as being unpatentable over the combination of U.S. Patent No. 6,087,543 (hereinafter the '543 patent) and U.S. Patent No. 6,166,273 (hereinafter the '273 patent). Applicants respectfully traverse this ground of rejection. The '543 patent is directed to a method of synthesizing fluorinated benzene from chlorobenzene and a metal fluoride composition at process temperatures from 175 °C to 450 °C. The '273 patent is directed to a method of synthesizing fluorinated benzenes and pyridines from benzenes and pyridines and a metal fluoride composition at process temperature between 250 °C and 550 °C.

As an initial matter, Applicants submit that the cited combination of references fails to raise a *prima facie* case of obviousness because there is no motivation to combine the cited references nor is there a reasonable expectation of success in arriving at the presently claimed invention in view of the teachings of the references. The basis for combinability as articulated by the Federal Circuit includes "some reason, suggestion, or motivation found in the prior art whereby a person of ordinary skill in the field of the invention would make the combination" (*In re Octiker*, 977 F.2d 1443, 1447 (Fed. Cir. 1992)). The Office Action indicates that motivation to combine the two references lies in higher temperature processes taught in the '273 patent. However, the high temperature processes of the '273 patent fail to realize a high yield. In fact, the high temperature processes of the '273 patent result in yields lower than those in the '543 patent. Thus, a skilled artisan would lack motivation to combine the references because modifying the processes of the '543 patent with the teachings of the '273 patent would not result in improved yields of monofluorobenzene.

Therefore, a skilled artisan reading the '543 patent in view of the '273 patent would not have had a reasonable expectation of successfully producing monofluorobenzene at temperatures such as those recited within the claims. Applicants also respectfully submit that one skilled in the art would not have had a reasonable expectation of synthesizing monofluorobenzene in view of the teachings of the cited references. As noted above, the high temperature processes of the '273 patent fail to realize a high yield. In fact, the high temperature processes of the '273 patent result in yields lower than those in the '543 patent.

The Office Action also indicates that motivation lies in applying the processes of the '543 patent to a broader class of starting materials. Applicants respectfully disagree with the asserted motivation. As noted above, the methods of the '273 patent result in low yields of fluorobenzene, even at high process temperatures. These low yields do not suggest that utilizing benzenes and pyridines in addition to chlorobenzenes would result in an improved synthesis. In contrast, the skilled artisan would likely conclude that broadening the aromatic reactants of the '543 patent to include non-chlorinated aromatics would yield a significant loss of product.

Moreover, even if the cited references are properly combinable, the claimed methods demonstrate unexpected results and superior yields over the processes of the cited references. The Federal Circuit has expressed the importance of secondary evidence, including unexpected results, in refuting obvious rejections.

[E]vidence of secondary considerations may often be the most probative and cogent evidence in the record. It may often establish that an invention appearing to have been obvious in light of prior art was not. It is to be considered as part of all the evidence, not just when the decisionmaker remains in doubt after reviewing the art.

*Stratoflex, Inc. v. Aeroquip Corp.*, 713 F.2d 1530, 1538-39 (Fed. Cir. 1983).

The methods of the claimed invention show a superior yield of monofluoroaromatics over the methods of the cited references. Evidence of surprisingly superior yields produced from aromatics is provided in Tables 1 and 2 on page 6 of the specification. The claimed methods produce yields of monofluorinated compounds higher than the conversions of the '273 patent regardless of the temperature or the ratio of the fluorinating agents and supports. The applicants submit that the '273

patent provides a method that barely converts benzene to fluorobenzene, as shown in its Table 1 (column 3), at temperatures even higher than that of the claimed methods. The '273's highest yield is merely 15% at a temperature of 550°C (see Table 2). In contrast, Table 2 in the subject application illustrates that the claimed methods produced higher yields (23.8% to 63.1%) at temperatures far less than 550°C.

Furthermore, as illustrated in Table 3 (page 7), the claimed methods also efficiently produce superior conversion percentages even over time. The smallest conversion to fluorobenzene in a spent bed is still more than double the highest conversion shown in the '273 patent (15%) (see Table 1).

Similarly, when the starting reagent is a chloroaromatic, the claimed methods still show superior conversion rates as compared to the conversion rates of the '543 patent. Example 3 (at pages 8-9) illustrates that the conversion rates are far superior to those in the cited references. For example, Table 6 shows that at 450°C, the fluorobenzene conversion rate of the claimed methods is over 10% higher than the methods of the '543 patent.

Applicants further note the yield of monofluorinated benzene decreases as the temperature is increased above 400°C in the '543 patent. As illustrated in Table 2, the yield of monofluorinated benzene peaks at a temperature of 400°C and decreases from 39% to 29% at a temperature of 450°C. Thus, the subject invention produces unexpectedly superior yields of monofluorobenzene as compared to the '543 patent.

Thus, Applicants submit that these unexpectedly superior results render the claimed invention unobvious over the cited references; accordingly, Applicants request reconsideration and withdrawal of the 35 U.S.C. § 103(a) rejection.

In view of the foregoing remarks and amendments to the claims, Applicants believe that the currently pending claims are in condition for allowance, and such action is respectfully requested.

APR 27 2006

The Commissioner is hereby authorized to charge any fees under 37 CFR §§1.16 or 1.17 as required by this paper to Deposit Account No. 19-0065.

Applicants invite the Examiner to call the undersigned if clarification is needed on any of this response, or if the Examiner believes a telephonic interview would expedite the prosecution of the subject application to completion.

Respectfully submitted,



Jenna M. Morrison

Patent Attorney

Registration No. 55,468

Phone No.: 352-375-8100

Fax No.: 352-372-5800

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A Professional Association  
P.O. Box 142950  
Gainesville, FL 32614-2950

JMM/ssa

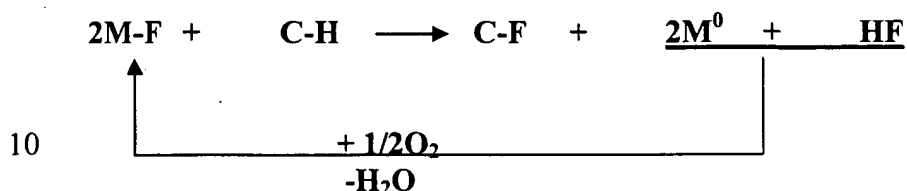
APR 27 2006



(a) in the presence of HF to regenerate a metal fluoride composition comprising  $\text{CuF}_2$  and/or  $\text{AgF}$ ; and (c) employing regenerated metal fluoride composition of (b) in (a).

Oxidative fluorination of aromatic compounds using transition metal fluorides is schematically represented below.

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With the simple metal fluorides, the fluorinating power depends on the redox potentials of the metal ions involved. Fluorides of the metal ions with  $E^0 > 1$  are very strong fluorinating agents giving rise to saturated products. Fluorides of the metal ions with  $E^0 < 0$  are inert towards aromatics. On the other hand, fluorides of the metal ions with  $1 > E^0 > 0$  are mild fluorinating agents. Metal fluorides useful as mild fluorinating agents in the present invention include but are not limited to  $\text{CuF}_2$ ,  $\text{AgF}$ ,  $\text{HgF}_2$ , and  $\text{Hg}_2\text{F}_2$ .

20

As illustrated in the examples attached hereto, a 73% conversion of benzene to fluorobenzene and difluorobenzene (ratio of fluorobenzene to difluorobenzene = 88:12) was obtained when benzene was contacted with a mixture of  $\text{CuF}_2$  and  $\text{AlF}_3$  ( $\text{CuF}_2:\text{AlF}_3$  ratio = 1:2) at a temperature of  $500^\circ\text{C}$ . At temperatures of  $450^\circ\text{C}$ , 44.3% conversion of benzene to fluorobenzene and difluorobenzene was observed (with a fluorobenzene:difluorobenzene ratio of 91:9). At  $400^\circ\text{C}$ , fluorobenzene was formed selectively with a yield of 24%. The use of  $\text{CaF}_2$  or  $\text{MgF}_2$  provided similar results to the use of  $\text{AlF}_3$ .

30

As is also illustrated in the examples, chlorobenzene can be converted to a mixture of fluorobenzene, difluorobenzene, and chlorofluorobenzene when reacted with mixtures of  $\text{CuF}_2$  and  $\text{AlF}_3$  (ratio of 1:2). At temperatures of  $500^\circ\text{C}$ , chlorobenzene is converted to fluorobenzene, difluorobenzene, and chlorofluorobenzene at a ratio of 85:7:9 (with a yield of 65%).

Additionally, we have found that ortho-, meta-, and para-chlorotoluenes can be converted to mixtures of fluorotoluenes when reacted with AgF at temperatures of at least about 350°C with conversion yields of 29%, 47%, and 57% respectively. Ortho- and meta-dichlorobenzenes are converted to meta-difluorobenzene (12% and 20% conversion respectively) plus chlorofluorobenzene (6.7% and 14.5% respectively). All chlorofluorobenzenes exhibited conversion to one product (meta-fluorobenzene) under the reaction conditions.

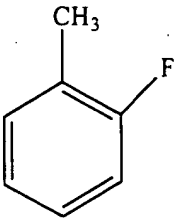
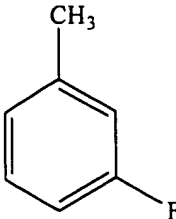
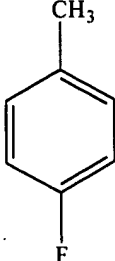
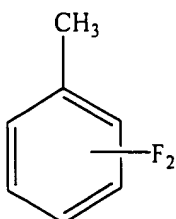
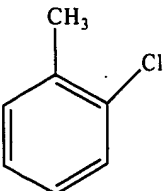
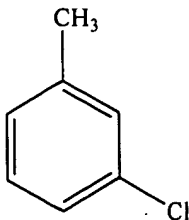
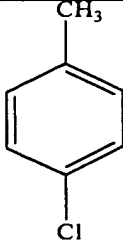
Following are examples that illustrate procedures for practicing the invention. These examples should not be construed as limiting. All percentages are by weight and all solvent mixture proportions are by volume unless otherwise noted.

Example 1: Fluorination of Benzene Utilizing  $\text{AlF}_3$  and  $\text{CuF}_2$  as the Fluorinating Agent

Anhydrous  $\text{AlF}_3$  (ALFAAESAR, 99.5%) and anhydrous  $\text{CuF}_2$  (ALFAAESAR, 99.5%) were used. To a weighed amount of  $\text{CuF}_2$ ,  $\text{AlF}_3$  (~35 mesh size) was mixed in different ratios (1:1, 1:1.5, 1:2, 1:2.5, 1:3). In a typical fluorination experiment, the metal fluoride mixture was loaded into a hastelloy reactor tube in a dry box. The reactor tube was heated to 500° C under a flow of Ar gas. The flow rate of the carrier gas was adjusted to 25 mL/min. Vaporized benzene was passed over the heated fluoride mixture. The duration of the reaction was about 1½ to 2½ hrs. At the end of the reaction, the reactor tube was swept out with the carrier gas. The organic product was analyzed using HP 6890 GC/ 5973 Mass Spectrometer. The inorganic residue was analyzed by powder X- ray diffractometer (XRD PHILIPS APD 3720).

Percentage conversions of benzene to the fluorinated products, *m*-fluorobenzene, fluorobenzene, and *o*-fluorobenzene, where the amount of  $\text{AlF}_3$  increases and  $\text{CuF}_2$  remains constant are given in Table 1.

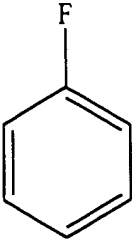
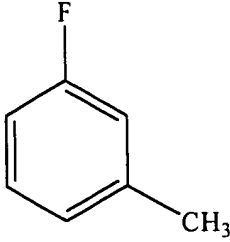
Table 8

Reactants	Temp. °C	Product Yields %			
					
 + AgF (3.5 g)	350	13.9	14.4	0.5	3.3
 + AgF (3.5 g)	350	6.7	25.7	15	7.7
 + AgF (3.5 g)	350	0.6	35.8	20.8	3.9

5

Attempts to fluorinate the resulting fluoroaromatic products listed in Table 1 yielded only a small increase in total fluoronation. The increased fluorination is shown in Table 9 for fluorobenzene and fluortoluene.

Table 9

Reactants	Temperature °C	Product Yields
	350	Only 2-4% fluorination
	350	

#### 5 Fluorination of dichlorobenzenes

All three dichlorobenzenes were fluorinated using the methods disclosed in Example 1. The fluorinating agent utilized was AgF and the four resulting product yields are shown in Table 10.

APR 27 2006

UNITED STATES PATENT AND TRADEMARK OFFICE

CERTIFICATE OF CORRECTION

PATENT NO. : 7,012,165

Page 1 of 2

APPLICATION NO.: 10/698,110

DATED : March 14, 2006

INVENTOR : William R. Dolbier, Jr., Buvaneswari Gopal

It is certified that errors appear in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

Title page,

Item (74), "& Salinwanchik" should read --& Saliwanchik--.

Column 3,

Line 20, " $\text{Hg}_2\text{F}_2$ . As illustrated" should read  
-- $\text{Hg}_2\text{F}_2$ .

As illustrated--.

Column 3,

Line 29, "fluorbenzene: diflorobenzene" should read --fluorobenzene: diflorobenzene--.

Column 4,

Line 41, "flurobenzene" should read --fluorobenzene--.

Column 11,

Line 1, "flourinate" should read --fluorinate--.

Line 3, "fluoronation" should read --fluorination--.

Line 4, "flourobenzene" should read --fluorobenzene--.

Line 4, "flurotoluene" should read --fluorotoluene--.

MAILING ADDRESS OF SENDER:

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APR 27 2006

# UNITED STATES PATENT AND TRADEMARK OFFICE

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PATENT NO. : 7,012,165

Page 2 of 2

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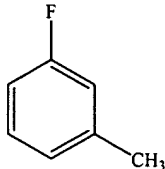
INVENTOR : William R. Dolbier, Jr., Buvanewari Gopal

It is certified that errors appear in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

### Column 12,

Lines 5-10, Table 9,

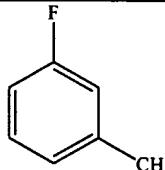
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Reactants	Temperature °C	Product Yields
	350	

”

should read

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### Column 15,

Line 3, “components a temperature” should read --components at a temperature--.

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APR 25 2006

# UNITED STATES PATENT AND TRADEMARK OFFICE

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PATENT NO. : 7,012,165

Page 2 of 2

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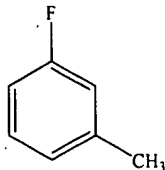
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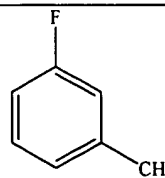
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